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We study SHG from metallo-dielectric multilayered structures with particular attention to the role played in the strong enhancement of generation process by the geometry of the elementary cell and by the excitation of short-range/long-range plasmons

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Plasmonic Second Harmonic Generation (SHG) from Metallo-Dielectric Multilayered Structures

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Abstract: We study SHG from metallo-dielectric multilayered structures with particular attention to the role played in the strong enhancement of generation process by the geometry of the elementary cell and by the excitation of short-range/long-range plasmons

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1. Introduction

SHG from metal surfaces in the non-plasmonic regime has been studied since the beginning of nonlinear optics [0] in the 60s. Nevertheless the subject of plasmonic SHG was addressed just several years later in 1974 [2] when SH from a single layer of silver in the Kretschmann geometry was studied both experimentally and theoretically. In this paper we analyze plasmonic SHG from metal/dielectric periodic stratifications. In particular we study SHG from structures made of Ag (silver) and MgF_2 (magnesium-fluoride) due to the particular simple procedure to grow these materials with standard sputtering or thermal evaporation techniques.

2. Results and Discussion

Let us start our analysis by describing in Fig.1 the types of geometries studied.

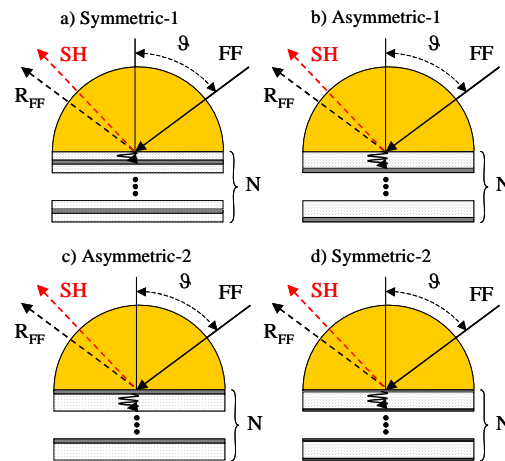Fig. 1. MgF_2/Ag multilayered structures are grown on a hemi-cylindrical prism. The output medium is air. a) Symmetric elementary cell (MgF_2/Ag/MgF_2). b) Asymmetric (MgF_2/Ag). c) Asymmetric (Ag/MgF_2). d) Symmetric (Ag/MgF_2/Ag). In the figures are also represented the input fundamental frequency (FF) pump field (λ=800nm, incident angle ϑ) and the reflected FF and SH fields.

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The calculations are based on the theoretical model that we have developed in Ref. [3]. Under TM polarization, the Helmholtz equation for the H-field at the SH can be written as follows (see Ref. [3] for more details):

$$\frac{d^2 H_{2\omega}}{dz^2} + \frac{4\omega^2}{c^2}\left(n_{2\omega}^2(z) - n_{in}^2 \sin^2\vartheta\right)H_{2\omega} = \frac{4\omega^2 n_{in}}{c}\sin\vartheta\left(\varepsilon_0 d_s^{(2)}\sum_k \delta(z - z_k)E_{z,\omega}^2\right) \quad . \quad (1)$$

In Eq.(1) we have neglected the volume quadratic nonlinearity of metals (Lorentz term) [3] and just considered their surface nonlinearity ($d_s^{(2)} = 10^{-18} m^2/V$) because in the plasmonic regime it is the only one that can be efficiently excited. The results of the conversion efficiency are summarized in Table 1.

Symmetric-1					Asymmetric-1			
N	d_{Ag} (nm)	d_{MgF2} (nm)	ϑ (^{0})	η	N	d_{Ag} (nm)	d_{MgF2} (nm)	ϑ (^{0})	η
1	34	342	80.6	5.96E-9	1	160	117	81.2	2.28E-9
2	28	209	82.1	1.11E-8	2	25	173	73.1	1.15E-8
3	22	155	82.5	2.04E-8	3	20	109	79.0	1.93E-8
4	18	123	82.7	2.32E-8	4	16	86	81.5	3.15E-8
5	15	101	82.8	4.90E-8	5	13	73	82.2	4.61E-8
6	13	86	83	6.87E-8	6	12	66	82.7	6.36E-8
7	12	77	82.8	9.08E-8	7	10	58	82.7	8.56E-8
8	10	66	83	1.17E-7	8	9	52	83.2	1.11E-7

Asymmetric-2					Symmetric-2			
N	d_{Ag} (nm)	d_{MgF2} (nm)	ϑ (^{0})	η	N	d_{Ag} (nm)	d_{MgF2} (nm)	ϑ (^{0})	η
1	34	600	67.3	4.12E-8	1	29	600	66.1	1.93E-8
2	26	600	67.6	3.23E-8	2	46	259	78	2.80E-8
3	25	600	67.7	3.18E-8	3	23	160	82.5	4.50E-8
4	24	600	67.7	3.18E-8	4	18	125	82.7	6.40E-8
5	20	84	63.4	4.10E-8	5	15	102	82.8	8.53E-8
6	17	119	81.9	6.80E-8	6	14	91	82.4	1.07E-7
7	15	103	81.7	9.78E-8	7	12	78	82.5	1.36E-7
8	13	89	81.9	1.31E-7	8	10	66	83	1.68E-7

Tab. 1. Configuration at which the maximum conversion efficiency (η) of the reflected SH at λ=400nm is reached. N is the number of elementary cells. d_{Ag} and d_{MgF2} are the total thickness of the materials in the elementary cell. The input intensity is 6GW/cm^2.

These results show that the conversion efficiencies can be up to three orders of magnitude greater than the conversion efficiencies experimentally found and theoretically predicted in Ref [3] in the non-plasmonic regime. Also from Table 1 one realizes that there are three angles at which the maximum conversion efficiencies are achieved: $\vartheta \sim 67^0$, $\vartheta \sim 63^0$, $\vartheta \sim 83^0$ which correspond respectively to the excitation of single interface plasmons ($\vartheta \sim 67^0$), long-range ($\vartheta \sim 63^0$)/short-range ($\vartheta \sim 83^0$) plasmons.

3. Conclusions

We have studied SH generation in the plasmonic regime for Ag/MgF$_2$ structures and found three orders of magnitude enhancement with respect to the non-plasmonic regime. Finally we would like to point out that our calculations also show that there are still vast margins of improvement for the conversion efficiencies by considering more sophisticated, non-periodic structures and by carefully choosing the output medium.

4. References

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